Combined statistical and dynamical model of ternary nuclear fission

J. P. Lestone

Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA (Received 27 October 2003; published 20 August 2004)

The statistical theory of particle evaporation from hot compound nuclei can be used to calculate the probability that particles are evaporated from the nuclear surface with not enough energy to surmount the Coulomb barrier. These quasievaporated particles exist between the nuclear surface and the Coulomb barrier for a short period of time before returning to the nuclear fluid. Occasionally, a quasievaporated charged particle emitted into the region surrounding the pre-scission neck material, fails to be reabsorbed by either of the main fragments as they accelerate away from each other after scission. This new particle emission mechanism can be used to explain many of the properties of ternary nuclear fission.

DOI: 10.1103/PhysRevC.70.021601 PACS number(s): 24.75.1i, 24.60.Dr, 25.85.Ca, 25.85.Ec

Ternary nuclear fission is a process by which a third charged fragment is generated in the fission process close to the plane perpendicular to the direction of the two main charged fragments. Extensive measurements on the properties of ternary fission have been made and substantially documented [1–4]. The energy spectra and angular distributions of these particles show that they are generated close to the scission (rupture) point, between the two main fission fragments. The dominant ternary-fission particles are alpha particles, which occur a few times per thousand fissions in thermal-neutron-induced fission of actinides (see Fig. 1).

The consensus has been that ternary fission is not associated with an evaporation (statistical) process. Several arguments have been used to support this consensus. The main argument is the high-energy cost for emitting ternary particles. The energy cost for ternary alpha particles has been calculated to be \sim 20 MeV [1]. This large energy cost for ternary fission, and the nuclear temperature of \sim 1 MeV for low-excitation-energy induced-fission scission configurations, leads to calculated ternary alpha-emission probabilities in a purely statistical model that are orders of magnitude lower than those experimentally determined [4]. Based on this, and other arguments, many have concluded that the energy required to produce ternary fission comes purely from a dynamical process. Many dynamical models of ternary fission exist [4–8].

More recently, γ - γ - γ coincidences and γ - γ -light charged particle coincidences have provided evidence for two different mechanisms for ternary fission, one hot and the other cold $[9-11]$. In alpha-particle-accompanied fission of ²⁵²Cf the majority of the fissions are hot [9], while for ¹⁰Be-accompanied fission the cold process appears to dominate [11]. The coolness of the fission fragments following beryllium-accompanied fission suggests that berylliumaccompanied fission cannot be an evaporative process.

Halpern [12] made a detailed attempt to describe the ternary-fission emission mechanism. He suggested that the energy required to emit ternary-fission particles is transferred to these particles via the sudden collapse of the neck stubs into the main fragments after scission. A sudden neck collapse is also invoked in the model presented here. The main difference between the suggestion of Halpern and the model presented here is that the potential ternary-fission particles in

our model are first produced in the neck rupture region via a statistical process. Our model can be considered a blending of two models similar, but not identical, to those proposed by Halpern [12] and Fong [13,14]. The statistical particle generation mechanism does not need to produce the full energy required for ternary fission. Instead, the statistical process only needs to produce light particles in the region of the neck rupture. Statistically generated particles that are between the main fragments and sufficiently far from the scission axis can fail to be reabsorbed by either of the retracting neck stubs, and can find themselves outside the Coulomb barrier of the post-scission configuration.

Calculating the properties of ternary fission within the framework of the proposed combined statistical and dynamical model involves many steps. First, the shape, temperature, and kinetic energy of nuclear-fission scission configurations need to be determined. Second, the properties of statistically generated particles beyond the nuclear surface need to be determined. Third, statistically generated particles that are not reabsorbed by the retracting neck stubs after scission must be identified. Finally, trajectory calculations are required to propagate the three fragments to their final states. The degree of complexity that could be incorporated into each one of these steps is, of course, high. In this article, an attempt is made to use very simple computational methods while preserving enough complexity for the calculations presented here to demonstrate the essentials of the proposed ternary-fission mechanism.

FIG. 1. Ternary-fission probabilities for $^{235}U(n_{th}f)$ [4].

The Langevin equation [15] is used, in conjunction with a one-dimensional (1D) finite-range-corrected nuclear potential energy as a function of the separation of the nascent fragment centers [16], to calculate the descent of fissioning nuclei to the scission point. The combination of a reducednuclear-viscosity coefficient of $\beta=1.5\times10^{21}$ s⁻¹, and a distance between mass centers at scission of R_{mc} ~ 2.6 times the nuclear radius of the corresponding spherical system (R_0) , leads to calculated final fragment kinetic energies in the fission of heavy elements that are in agreement with the Viola systematics [17]. The radii of spherical nuclei are assumed to be $1.22 \times A^{1/3}$ fm. This is consistent with the sharp-surface Coulomb energy used in the nuclear potential-energy calculations [16]. The chosen reduced-nuclear-viscosity coefficient is consistent with the average viscosity at large deformation, calculated using the surface-plus-window dissipation mechanism [18]. The Langevin calculations for thermalneutron-induced fission of ^{235}U give a mean nuclear temperature at scission of *T*=1.03 MeV with a standard deviation of 0.05 MeV (assuming a Fermi-gas level-density parameter, $a = A/8.6 \text{ MeV}^{-1}$). The corresponding mean kinetic energy of the fragments at scission is 15 MeV with a standard deviation of 3 MeV. The temperature at scission and the kinetic energy of the fragments at scission are correlated, because the total energy of the system is conserved. The calculated standard deviation of the fragment kinetic energy is lower than the measured value, because the present calculation is a simple 1D calculation with a fixed fragment mass ratio and fixed distance between mass centers at scission.

Using the standard statistical theory of particle evaporation from hot compound nuclei [19], one can show that in the classical limit, the particle emission rate per unit area from nonaccelerating hot nuclear matter is

$$
R = \frac{(2s+1)\mu T^2}{4\pi^2 \hbar^3} \exp(-\Delta E/T),\tag{1}
$$

where *s* is the spin of the evaporated particle, μ is the reduced mass of the particle-daughter system, and ΔE is the minimum energy cost. ΔE is the particle binding energy plus the height of the corresponding Coulomb emission barrier. Many more particles than those successfully evaporated from hot nuclear matter, attempt to escape the nuclear fluid but are returned to the fluid by the strong nuclear force. These statistically generated particles exist between the nuclear surface and the Coulomb barrier. The probabilities of finding these quasievaporated particles as a function of height above the nuclear surface, and the velocity distributions of these particles, are predicted by statistical physics. In the limit of a shaped-surface nuclear fluid, the probability that a given particle type exists above a unit area of surface, between the nuclear surface and the Coulomb barrier, is given by

$$
P = \frac{\sqrt{2\pi}(2s+1)\mu^{3/2}T^{5/2}}{4\pi^2\hbar^3F_S} \exp(-\Delta E_S/T),
$$
 (2)

where ΔE_S is the minimum energy cost for getting the particle to the surface in question and F_S is the force on the particle as it crosses this surface.

J. P. LESTONE PHYSICAL REVIEW C **70**, 021601(R) (2004)

FIG. 2. A scission configuration. The thicker curve shows the shape of a nucleus just before the neck snaps. The semicircles show the location of spherical fragments immediately following scission, assuming an instantaneous retraction of the neck stubs. The small solid circles highlight locations on the nuclear surface that do not undergo high acceleration during the collapse of the neck stubs. The dashed parabola represents the surface beyond which particles fail to be reabsorbed by the retracting neck stubs.

To be classically evaporated from a nonaccelerating nuclear fluid, a charged particle must have enough energy to reach a point above the nuclear surface, where the Coulomb repulsion and the nuclear attraction are equal. This height is $\sim r_p + 3\delta$ [20] for nearly spherical nuclei, where r_p is the radius of the particle and δ is the diffuseness of the Woods-Saxon nuclear potential. However, if the nuclear fluid is accelerating away from a particle attempting to escape, then the height that must be reached is lower. Figure 2 shows some key features of a scission configuration. The parabola in Fig. 2 represents a hyperbolical surface beyond which it is assumed that particles fail to be reabsorbed by the retracting neck stubs. A definitive calculation of this surface would be very complex. Here, this surface is defined using three rings around the neck region. Two of these are a distance of r_p $+3\delta$ along the normals to the scission configuration from the small circles shown in Fig. 2, and the third is $r_p + \Delta_p$ above the neck rupture location. All statistically generated particles beyond this surface are assumed to be unaffected by the collapse of the neck stubs, while all particles below are assumed to be reabsorbed. The quantity Δ_p is the only free parameter, and is adjusted to reproduce only the measured ternaryfission probabilities. If the mechanism proposed here is responsible for ternary fission, then we might expect Δ_p to vary smoothly with the mass and charge of the ternary-fission light particles.

The probability of generating a potential ternary-fission particle, as a function of both Δ_p and the scission temperature *T*, is obtained by integrating Eq. (2) over the hyperbolical surface represented by the dashed curve in Fig. 2. The final result is obtained by averaging over the ensemble of scission temperatures generated by the Langevin calculations described above. The only term in Eq. (2) that is difficult to calculate is the energy cost, ΔE_S . This is determined by the difference in the nonthermal energy of the scission configuration with no statistically generated particles, and the nonthermal energy of the scission configuration with the required number and type of nucleons removed to form a single particle at the surface in question, and can be expressed as

$$
\Delta E_S = M_D + M_{LP} + V_C + V_N - M_P, \tag{3}
$$

where M_D and M_P are the $T=0$ masses of the daughter and parent systems, respectively, and are estimated using the modified liquid drop model [16]. M_{LP} is the measured ground-state mass of the ternary-fission light particle. V_C and V_N are the Coulomb and nuclear potential energies, respectively, of the light particle in the field of the daughter system. The shape of the parent system is the same as the scission shape in the Langevin calculations. The deformation of the daughter was determined by assuming that the centers of mass of both halves of the scission configuration remain fixed during the statistical generation of the ternary-fission light particle. The Coulomb potential energy, V_C , is obtained assuming a point particle in the field of the sharp-surfaced daughter system.

It is well established that the depth of the Woods-Saxon potential for the interaction of a single nucleon with a nucleus is \sim 50 MeV, with a diffuseness close to 0.6 fm [21]. Much deeper depths have been assigned to deuteron, 3 He, and triton potentials. However, to model the statistical evaporation of alpha particles from hot nearly spherical nuclei, many statistical model codes (e.g., [22,23]) use an alpha-particle potential with a depth of 50 MeV [24]. The depths of the real part of the Woods-Saxon potentials, determined by the elastic scattering of heavy ions, vary from 10 to 70 MeV [25]. By constraining the diffuseness parameters, the extracted depths appear to be more clustered about 50 MeV for heavy ions with $10 < A < 20$ [25]. It should be noted that the elastic-scattering data are more sensitive to the outer regions of the nuclear potential, and thus these potential depths may contain considerable uncertainty. The true *A* and *Z* dependence of the nuclear potential around the neck region is made more complex by the high surface curvature around the neck, and the inverted curvature along the neck. Given the uncertainty associated with defining the nuclear potential energy of a particle in the region of the neck, a simple model is used here. V_N is estimated using a Woods-Saxon potential with a maximum depth of 50 MeV, and a diffuseness of δ =0.6 fm, independent of mass and charge of the light particles. The Woods-Saxon potential is assumed to be half its maximum depth when the center of mass of the particle is separated from the nuclear surface by the radius of the light particle.

To define the initial conditions of the three charged particles in ternary fission, it is assumed that the nascent fragments instantaneously collapse into spherical fragments at the moment of scission. The initial velocity and position distributions of the light particles are completely determined by statistical physics. The probability of generating a ternaryfission particle is a strong function of the distance of closest approach of the dashed curve shown in Fig. 2 and the nuclear surface at scission. This is controlled by the free parameter Δ_p . The initial velocity distribution of the light particle is a function of the nuclear temperature at scission. Due to the strong dependence of ΔE_S on the distance from the nuclear surface, the initial positions of the light particles are on and just above the hyperbolical surface represented by the dashed curve in Fig. 2, and strongly peaked about the location of closest approach of the hyperbolical surface and the nuclear surface at scission. Three-body trajectory calculations are used to propagate the two main spherical fragments and the generated ternary-fission light particles in time. The three particles each interact via Coulomb fields and Woods-Saxon

FIG. 3. The values for Δ_p needed to reproduce ²³⁵U(n_{th} *f)* ternary-fission probabilities [4]. The line is only to guide the eye.

potentials with depths and diffusenesses as given above. Some fraction of the initial starting points do not lead to ternary fission, because the light particle is reabsorbed by one of the spherical fragments. The final calculated properties of the ternary-fission particles are obtained by an average over the ensemble of correlated scission temperatures and the fragment kinetic energies, as generated by the Langevin calculations described above.

Assuming only symmetric fission in the three-body trajectory calculations leads to angular distributions for the light particles that peak symmetrically between the main fragments and gives widths for the corresponding angular distributions that are less than the measured values. Using a realistic mass distribution for the main fragments in the threebody trajectory calculations rectifies these deficiencies in the angular distributions.

Using the approach described above, the probability of ternary fission, and the properties of the corresponding light particles, can be calculated for various fission reactions as a function of both particle type and the single free parameter Δ_p . For the ²³⁵U($n_{\text{th}}f$) ternary-fission reaction, the free parameter Δ_p was adjusted to reproduce each of the measured emission probabilities of light particles from protons to 12 Be. These results are shown in Fig. 3. The Δ_p parameter has a strikingly linear dependence on the empirical scaling parameter $A+3(Z-1)/2$ for ternary fission involving helium, lithium, and beryllium isotopes. The dependence of Δ_p on *A*

FIG. 4. Measured [26] and calculated (solid curves) energy spectra for ⁴He, ⁶He, and ³H in ²³⁵U(n_{th} ,*f*) ternary fission. The disagreement on the low-energy side of the ⁴He spectrum is due to the breakup of ⁵He emission into ⁴He plus a neutron. The data and curves are normalized to have the same maximum height.

FIG. 5. Measured [4] and calculated (solid curves) mean energies (a), and the full width at half maximum (FWHM) of the energy spectra (b) of the light particles in $^{235}U(n_{th}f)$ ternary fission.

and *Z* is very sensitive to the assumed nature of the interaction potential between the light particles and the neck material. For example, the apparent strong dependence of Δ_p on mass can be reduced by introducing an increasing depth of the Woods-Saxon potential with increasing mass of the light particle. No great significance should be inferred from the details of the mass and charge scaling of Δ_p shown in Fig. 3. What is of importance is the fact that a smooth relationship exists.

Figure 4 compares measured and calculated ⁴He, ⁶He, and ³H energy spectra in ²³⁵U(n_{th} f) ternary fission. The agreement between the model calculations and experiment is impressive because no free parameters are adjusted to obtain the agreement shown in Fig. 4. Figure 5 compares measured and calculated mean energies and the full width at half maximum (FWHM) of the energy spectra of light particles from $^{235}U(n_{\text{th}}f)$ ternary fission. The model overpredicts the mean energy of the protons, deuterons, and the lithium and beryllium isotopes. The predicted mean energies of the light particles are sensitive to the initial velocity distribution of the light particles, the scission configuration neck radius, the assumed distance between mass centers, and the kinetic energy of the two main fragments at scission. The widths of the light-particle energy spectra provide a more critical test of the proposed ternary fission production mechanism, because these widths are sensitive to the assumed initial velocity distribution of the light particles, and less sensitive to other assumed properties of the scission configuration. The calculated widths are in good agreement with the experimental data.

J. P. LESTONE PHYSICAL REVIEW C **70**, 021601(R) (2004)

Given the experimental evidence that suggests berylliumaccompanied ternary fission is a cool process [10,11], it is difficult to understand why the ternary-fission model proposed here appears to be so successful. However, some other recent works also seem to support a hot ternary-fission process. The nuclear temperature of scission configurations in spontaneous and low-energy neutron-induced ternary fission of heavy elements has been estimated to be \sim 1.1 MeV, using isotope thermometry [27]. Koparch *et al.* [28] have measured the ternary-fission production probability for the particle-unstable short-lived 2.26 MeV 3⁺ excited state of ${}^{8}Li$ (${}^{8}Li^{*}$) in spontaneous fission of ${}^{252}CF$. The ${}^{8}Li^{*}$ datum is a powerful test of the proposed ternary-fission model. This is because the ⁸Li^{*} result cannot be obtained by an interpolation or extrapolation of measured ground-state emission probabilities. In addition, the only quantities that are changed to calculate the ${}^{8}Li^{*}$ ternary-fission probability relative to the lithium ground-state emissions are an additional energy cost of 2.26 MeV and a change in ${}^{8}Li$ spin from 2 to 3. The model calculation for the ratio of the ⁸Li^{*} ternary-fission probability in the spontaneous fission of 252Cf to the probability summed over all lithium ground states is 5.3%, in agreement with the experimental result of $6\pm2\%$. This is further evidence that ternary-fission light particles are being produced in the neck region via a statistical process.

Another mechanism has been proposed for ternary nuclear fission. This mechanism involves a statistical (evaporation) process that produces quasievaporated particles that exist between the nuclear surface and the Coulomb barrier. The sudden snap of the neck stubs can cause a quasievaporated charged particle, high above the neck-rupture location, to switch from being inside to outside the Coulomb barrier. A key feature of the proposed mechanism is that the statistical process does not need to produce light charged particles that can escape the Coulomb barrier using energy derived solely from the heat bath. It must be emphasized that the emission mechanism described here is classical. The quasievaporated particles are in the classically allowed region between the nuclear surface and the emission barrier, and will return to the nuclear fluid if the field they experience is independent of time. However, a sudden rearrangement of the nuclear fluid can lead to the ejection of some quasievaporated particles.

Based on the experimental results that are understood within the framework of the proposed combined statistical and dynamical model of ternary fission, the consensus that ternary fission is not associated with an evaporation process should be reevaluated.

The author would like to thank P. Russo, A. J. Sierk, and R. Vandenbosch for their valuable input during the writing of this manuscript.

COMBINED STATISTICAL AND DYNAMICAL MODEL OF… PHYSICAL REVIEW C **70**, 021601(R) (2004)

- [1] I. Halpern, Annu. Rev. Nucl. Sci. **21**, 245 (1971).
- [2] R. Vandenbosch and J. R. Huizenga, *Nuclear Fission* (Academic, New York, 1973).
- [3] A. K. Sinka, D. M. Nadkarni, and G. K. Mehta, Pramana **33**, 85 (1989).
- [4] C. Wagemans, *The Nuclear Fission Process* (CRC, Boca Raton, 1991).
- [5] N. Carjan, J. Phys. (Paris) **37**, 1279 (1976).
- [6] V. A. Rubchenya and S. G. Yavshits, Z. Phys. A **329**, 217 (1988).
- [7] O. Tanimura and T. Fliessbach, Z. Phys. A **328**, 475 (1987).
- [8] N. Carjan, K. Weingaertner, and J. P. Theobald, Verh. Dtsch. Phys. Ges. **23**, C4.6 (1988).
- [9] A. V. Ramayya *et al.*, Phys. Rev. C **57**, 2370 (1998).
- [10] A. V. Ramayya *et al.*, Phys. Rev. Lett. **81**, 947 (1998).
- [11] A. V. Ramayya *et al.*, Prog. Part. Nucl. Phys. **46**, 221 (2001).
- [12] I. Halpern, in *Proceedings of the IAEA Symp. Phys. Chem. Fission, Salzburg 1965* (International Atomic Energy Agency, Vienna, 1965), Vol. 2, p. 369.
- [13] P. Fong, Phys. Rev. C **2**, 735 (1970).
-
- [14] P. Fong, Phys. Rev. C **3**, 2025 (1971).
- [15] Y. Abe, C. Gregoire, and H. Delagrange, J. Phys. Colloq. **47**, 329 (1986).
- [16] J. P. Lestone, Phys. Rev. C **51**, 580 (1995).
- [17] V. E. Viola, K. Kwiatkowski, and M. Walker, Phys. Rev. C **31**, 1550 (1985).
- [18] J. R. Nix and A. J. Sierk, in *Proceedings of the International School—Seminar on Heavy Ion Physics Dubna, USSR, 1986*, p. 453.
- [19] T. D. Thomas, Nucl. Phys. **53**, 558 (1964).
- [20] D. C. Williams and T. D. Thomas, Nucl. Phys. **A92**, 1 (1967).
- [21] C. M. Perey and F. G. Perey, At. Data Nucl. Data Tables **17**, 1 (1976).
- [22] A. Gavron, Phys. Rev. C **21**, 230 (1980).
- [23] J. P. Lestone, Phys. Rev. C **59**, 1540 (1999).
- [24] J. R. Huizenga and G. Igo, Nucl. Phys. **29**, 462 (1962).
- [25] M. Lozano and G. Madurga, Nucl. Phys. **A334**, 349 (1980).
- [26] P. D'hondt *et al.*, Nucl. Phys. **A346**, 461 (1980).
- [27] M. N. Andronenko *et al.*, Eur. Phys. J. A **12**, 185 (2001).
- [28] Y. N. Kopatch *et al.*, Phys. Rev. C **65**, 044614 (2002).